

THE PATENTS ACT, 1970

of Application and Provisional Specification filed on 9.5 2000 in respect of Patent Application No 423/MUM/2000 of Hindustan Lever Ltd, a company incorporated under the Indian Companies Act, 1913 and having its registered office at Hindustan Lever House, 165/166, Backbay Reclamation, Mumbai=400 020, Maharashtra India.

This certificate is issued under the powers vested on me under Section 147(1) of the Patents Act, 1970.......

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Dated this 20 H day of A/706

2001。

(N.K.Garg)
Asst Controller of Patents&Designs

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FORM 1

THE PATENTS ACT, 1970 (39 of 1970)

APPLICATION FOR GRANT OF A PATENTS [See Sections 5 (2), 7, 54 and 135 and rule 33A)



- 1. We, HINDUSTAN LEVER LIMITED, a company incorporated under the Indian Companies Act, 1913 and having its registered office at Hindustan Lever House, 165/166, Backbay Reclamation, Mumbai -400 020, Maharashtra, India
- 2. hereby declare -
 - •

(a) that we are in possession of an invention titled

SOIL RELEASE AGENTS FOR DETERGENT FORMULATIONS

- (b) that the Provisional specification relating to this invention is filed with this application.
- (c) that there is no lawful ground of objection to the grant of a patent to us.
- 3. further declare that the inventors for the said invention are

KUMAR Velayudhan Nair Gopa, 4B, Anusandhan, Hindustan Lever Research Centre Flats, I.C.T. Link Road, Chakala, Andheri East, Mumbai-400 099. Maharashtra, India, Indian Citizen, MOULEE Anuradha, 6A, Agresar, Hindustan Lever Research Centre Flats, I.C.T. Link Road, Chakala, Andheri East, Mumbai-400 099. Maharashtra, India, Indian Citizen

- 4. We, claim the priority from the applicant(s) filed in convention countries, particulars of which are as follows:-
- 5. We state that the said invention is an improvement in or modification of the invention, the particulars of which are as follows and of which we are the applicant/patentee.
- 6. We state that the application is divided out of our application, the particulars of which are given below and pray that this application deemed to have been filed on under Section 16 of the Act.
- 7. That we are the assignee of the true and first inventors
- 8. That our address for service in India is as follows:-

S.Majumdar & Co., 5 Harish Mukherjee Road, Calcutta - 700 025, India

423 mum 2000

Roentrod No 5000 in Employed

A23 = MUM 2000 Vido Bairo No 59/ in 100

Rochbor of Valuabion. Rochbor

G. F. Luck

C. F. Luck

9. Following declaration was given by the inventor(s) or applicant(s) in the convention country:

We, the true and first inventors for this invention or the applicant(s) in the convention country declare that the applicant(s) herein is/are our assignee or legal representative.

KUMAR Velayudhan Nair G pa, 4B, Anusandhan, Hindustan Lever Research Centre Flats, I.C.T. Link Road, Chakala, Andheri East, Mumbai-400 099. Maharashtra, India, Indian Citizen, MOULEE Anuradha, 6A, Agresar, Hindustan Lever Research Centre Flats, I.C.T. Link Road, Chakala, Andheri East, Mumbai-400 099. Maharashtra, India, Indian Citizen

Volay Whan Nor Copa Kunas KUMAR Velayudhan Nair Gopa

0 ...

MOULEE Anuradha

- 10. That to the best of our knowledge, information and belief the fact and matters stated herein are correct and that there is no lawful ground of objection to the grant of patent to us on this application.
- 11. Followings are the attachment with the application:
- (a) Provisional Specification (3 copies)
- (b) Drawings (3 copies)
- (c) Declaration of Inventorship Form-5
- (d) Fee Rs. 5000 in Cheque bearing No 244520 Dated 6/5/2000 drawn on ANZ Grindlays Bank, Mumbai

We request that a patent may be granted to us for the said invention.

Dated this 9th day of May 2000.

HINDUSTAN LEVER LIMITED.

To

The Controller of Patents The Patent Office Mumbai

FORM -2

THE PATENTS ACT, 1970 (39 of 1970)

PROVISIONAL SPECIFICATION

(See Section 10)

SOIL RELEASE AGENTS FOR DETERGENT FORMULATIONS

HINDUSTAN LEVER LIMITED, a company incorporated under the Indian Companies Act, 1913 and having its registered office at Hindustan Lever House, 165/166, Backbay Reclamation, Mumbai -400 020, Maharashtra, India

The following specification describes the nature of the invention:

- 9 MAY 2000

TECHNICAL FIELD:

The present invention relates to novel soil release polymers that are capable of acting on both cotton and polyester fabric or their blends for removal of oily as well particulate soils and a process for the preparation of the same. The novel soil release polymers are hydrophobic and anionic polysaccharides.

BACKGROUND OF THE INVENTION:

The washing of soiled fabrics with a laundry detergent is essentially a two step process. In the first stage, the detergent removes the soil particles from the fabric and suspends them in the soil solution. In the second stage the detergent must prevent soil particles and other insolubles from redepositing on the cloth before and after the fabric is removed from the soil solution or the rinse solution. Polymers are known to aid both processes – soil release polymers enhance soil removal from the fabric whilst anti-redeposition polymers prevent the deterged soil from depositing on the fabric. The thrust in recent times has been the development of soil release polymers (SRPs) which can be incorporated into detergent formulations to enhance the removal of soil from the fabric. SRPs adsorb on the fabric surface, modifying properties like hydrophilic/hydrophobic nature of the fabric and its surface energy. Consequent soil removal is greater than what is possible with a conventional detergent formulation.

Most soil release polymers disclosed in the literature are for removal of oily or fatty soils from polyester. Polyester is a hydrophobic fabric and removal of hydrophobic oily soils from the fabric has historically been a problem. The problem has been circumvented in part by using soil release polymers which have both hydrophobic and hydrophilic segments. These polymers adsorb strongly on the fabric, are easily dispersed or dissolved in a surfactant, are compatible with the components of the detergent formulation and show superior oily soil removal.

Various soil release polymers have been disclosed in literature for removal of oily soils from polyester. The common soil release polymers are polyesters that have been hydrophilically modified and these polymers are useful in the removal of oily soil from polyester fabrics. EP185427 discloses anionic modification of these polymers to increase hydrophilicity and improve water solubility of the SRP is known but they these are also useful only in the removal of oily soil from polyester fabrics.

GB2322137 (corresponding to our copending application 624/BOM/96) discloses the hydrophobic modification of starch (where starch is the hydrophilic segment) and its use as a soil release polymer, in particular for detergency of oily soil from polyester fabric. Hydrophobic modification was carried out by graft copolymerisation of hydrophobic monomers on starch.

US3948838 discloses the use of copolymers of hydrophobic acrylate monomers and water soluble monomers like acrylic acid, as oily soil release agents for cotton fabrics.

While the literature on soil release polymers is vast, the emphasis has been on the use of modified polyesters as SRPs and for enhancing removal of oily soil from polyester. However, soil release polymers that enhance removal of both oily and particulate soil from both cotton and polyester fabric are not reported.

It has now been found that anionically modified hydrophobic graft copolymers of polysaccharides can function as soil release polymers on both cotton and polyester for the removal of oily and particulate soils. These polymers can be formulated as detergent compositions or rinse conditioners. Thus the principal object of the present invention is to provide novel soil release polymers that are capable of acting on both cotton and polyester fabric or their blends for removal of oily as well particulate soils and a process for the preparation of the same.

It is a further object of this invention to formulate synergistic detergent compositions incorporating such soil release polymers for superior removal of oily and particulate soil from both cotton and polyester fabric or their blends.

DESCRIPTION OF THE INVENTION:

According to one aspect of the invention there is provided a polysaccharide based soil release polymer of representative formula I

$$R_1$$
-COOH/SO $_3$ H $\\$ -----(sugar)-----(sugar)------

Formula I

Where R is a long chain hydrophobic polymer and R_1 is an alkyl group. The sugar moiety may also be a substituted sugar.

Formula II (Fig. 1a-1d) are representative formulas of anionically modified hydrophobic graft copolymer of polysaccharides of the invention.

Such that $R_1 = -H$, $-CH_3$ and/or $-C_2H_5$

and $R_2 = -COOCH_3$, $-COOC_2H_5$, $-COOC_3H_7$, $-C_6H_5$ and/or $-OCOCH_3$

and R' = -CH₂COOH, -CH₂COO M^+ , -(CH₂-CHSO₃H-)_n or -(CH₂-CHSO₃M⁺-)_n where M is an alkali metal

According to another aspect of the invention there is provided a process for preparing a soil release polymer of formula II comprising the steps of

anionically modifying (by carboxylating or sulphonating) the polysaccharide (such polymers being represented by Fig. 2a-2d such that R' = -CH₂COOH, -CH₂COOM⁺, - (CH₂-CHSO₃H-)_n or -(CH₂-CHSO₃M⁺-)_n where M is an alkali metal), said polysaccharide having been optionally subjected to graft copolymerisation using a hydrophobic monomer to provide a hydrophobic graft copolymerised polysaccharide (such polymers being represented by Fig. 3a-3d where

$$R = -CH_2 - C - (-CH_2 - C -)_n - \\ | R_2 | R_2$$

where R_1 = -H, -CH₃ and/or -C₂H₅ and R_2 = -COOCH₃, -COOC₂H₅, -COOC₃H₇, -C₆H₅ and/or -OCOCH₃)

ii) graft copolymerising the anionically modified polysaccharide if not already copolymerised.

The step of graft copolymerisation of the polysaccharide using a hydrophobic monomer is carried out prior to or after the step of anionic modification. A preferred polysaccharide is starch and when polymer is produced by the aforesaid process using starch as the polysaccharide the polymer is made water soluble by heating at a temperature of 70-90°C in the presence of water and the polymer recovered therefrom by drying.

According to another aspect of the invention there is provided a fabric washing composition comprising

from 5-50% by wt. of detergent active;

from 1-50% by wt. of detergency builder; and

from 0.01-10% by wt. of the polymer of formula I.

According to a further aspect of the invention there is provided a fabric washing kit comprising an enclosure having a conventional detergent formulation; a second enclosure having the polymer of formula I; and instruction material containing directions for the use of the detergent formulation and the polymer during fabric washing.

DETAILED DESCRIPTION OF THE INVENTION:

The polymers of the invention have a polysaccharide backbone with multiple hydroxyl groups. Both graft copolymerisation and anionic modification of the polysaccharide can take place through the primary and secondary hydroxyl groups on the polysaccharide backbone. Graft copolymerisation can also be initiated by H abstraction from the sugar residue. The anionic groups (carboxyl and/or sulfonic groups) are distributed along the backbone of the polysaccharide. The hydrophobic modification is provided by hydrophobic polymers which are long chain polymers attached to the polysaccharide backbone.

The essential feature of the invention relates to novel soil release polymers of formula I that are anionically modified hydrophobic graft copolymers of polysaccharides and are capable of acting on both cotton and polyester fabric or their blends for removal of oily as well particulate soils and a process for the preparation of the same.

The polysaccharide which is the hydrophilic part of the molecule is chosen from cellulose, guar gum, starch, tamarind kernel powder and starch is particularly preferred. The source of the starch may be any known source of starch, for example, wheat, rice, oat, tapioca, maize, potato, arrowroot etc. Acid or enzymatically degraded starch or oxidised starch could also be used for chemical modification. The starch used may be in the native form or gelatinised form. The term gelatinisation refers to rupture of the starch granule at elevated temperatures in presence of water. When the polymer is prepared using native starch it is preferred to subject the same to a temperature of 70-90°C in presence of water to make the polymer water soluble.

The hydrophobic part of the molecule is a polymer of an ethylenic unsaturated monomers, preferably acrylic monomers. The structure of the acrylic monomer is as shown below. R₂ is an alkyl group (C1-C3), preferably methyl, and R₁ is H or C1-C2. The grafting is from 0.5-10% on starch. Grafting may also be carried out with other unsaturated, hydrophobic monomers such as styrene or vinyl acetate. Mixtures of these monomers may also be used.

$$COOR_2$$
 $|$
 $H_2C=C-R_1$



The polysaccharide has to be anionically modified and an graft copolymerised and it is not particularly relevant for the present invention as to which step is carried out first. Although it is particularly preferred to carry out the step of graft copolymerisation of the polysaccharide first followed by anionic modification.

The nature of the hydrophobic monomer and the hydrophilic polysaccharide has already been defined. The polysaccharide is graft copolymerised by contacting the redox initiators such as ferrous ammonium sulphate and hydrogen peroxide along with or without additional ascorbic acid or ceric ammonium nitrate and dilute nitric acid in an aqueous medium at a temperature range of 20-50°C followed by the addition of the hydrophobic monomer and subsequent polymerisation to yield the modified polysaccharide. It is preferable to use entrainers such as urea to facilitate the reaction. The level of the hydrophobic modification is from 0.1 to 10% by weight of the polysaccharide and the molecular weight of the polymer so formed is from 500-5,00,000.

The hydrophobic graft copolymer is preferably reacted with a carboxylating or sulphonating reagent selected from haloacetic acid, alkali metal salt of a haloacetic acid, vinyl sulphonic acid, the alkali metal salt of the vinyl sulphonic acid. It is preferred to use haloacetic acid to provide anionic groups on the polysaccharide. The level of carboxylation is from 0.1-10% by weight of

polysaccharide/modified polysaccharide, preferably 0.1-5%. The process of anionic modification may be carried out in presence of a solvent such as water/isopropanol mixtures or as a dry process in the absence of such solvents.

The soil release polymer of the invention and prepared by the above process may be used to treat fabric by incorporating it into detergent compositions using conventional methods or can be used as aqueous solutions to obtain the desired result of improved detergency obtained by enhanced soil removal from the fabric.

The polymer is incorporated at the level of 0.01-10% by weight of the detergent composition, preferably 0.5-5% by weight.

The detergent formulations of the invention, also contain, as in conventional formulations, detergent actives (surfactants) and builders and auxillaries. Auxillaries include sequestrants, dye-transfer inhibitors, perfumes, bleaches, enzymes, flourescers, optical brighteners, fungicides, germicides, hydrotropes etc.

The detergent composition may be in the form of a powder, bar, paste or liquid.

The detergent active is generally chosen from an anionic, nonionic, cationic, zwitterionic detergent active or mixtures thereof. 5-50% of the active can be incorporated in the formulation.

Anionic surfactants which can be used in the compositions of the invention are both soap and non-soap detergents compounds. Especially suitable anionic detergent active compounds are water soluble salts of organic sulphuric reaction products having in the molecular structure an alkyl radical containing from 8 to 22 carbon atoms, and a radical chosen from sulphonic acid or sulphur acid ester radicals and mixtures thereof.

The preferred water-soluble synthetic anionic detergent active compounds are the alkali metal (such as sodium and potassium) and alkaline earth metal (such as calcium and magnesium) salts of higher alkyl benzene sulphonates and mixtures with olefin sulphonates and higher alkyl sulphates, and the higher fatty acid monoglyceride sulphates. The most preferred anionic detergent active compounds are higher alkyl aromatic sulphonates such as higher alkyl benzene sulphonates containing from 6 to 20 carbon atoms in the alkyl group in a straight or branched chain, particular examples of which are sodium salts of higher alkyl benzene sulphonates or of higher-alkyl toluene, xylene or phenol sulphonates, alkyl naphthalene sulphonates, ammonium diamyl naphthalene sulphonate, and sodium dinonyl naphthalene sulphonate.

Suitable nonionic detergent active compounds can be broadly described as compounds produced by the condensation of alkylene oxide groups, which are hydrophilic in nature, with an organic hydrophobic compound which may be aliphatic or alkyl aromatic in nature. The length of the hydrophilic or polyoxyalkylene radical which is condensed with any particular hydrophobic group can be readily adjusted to yield a water-soluble compound having the desired degree of balance between hydrophilic and hydrophobic elements.

Particular examples include the condensation product of aliphatic alcohols having from 8 to 22 carbon atoms in either straight or branched chain configuration with ethylene oxide, such as a coconut oil ethylene oxide condensate having from 2 to 15 moles of ethylene oxide per mole of coconut alcohol; condensates of alkylphenols whose alkyl group contains from 6 to 12 carbon atoms with 5 to 25 moles of ethylene oxide per mole of alkylphenol; condensates of the reaction product of ethylenediamine and propylene oxide with ethylene oxide, the condensate containing from 40 to 80% of polyoxyethylene radicals by weight and having a molecular weight of from 5,000 to 11,000; tertiary amine oxides of structure R₃NO, where one group R is an alkyl group of 8 to 18 carbon atoms and the others are each methyl, ethyl or hydroxyethyl groups, for instance dimethyldodecylamine oxide; tertiary phosphine oxides of structure R₃PO, where one group R is an alkyl group of from 10 to 18 carbon atoms, and the others are each alkyl or hydroxyalkyl groups of 1 to 3 carbon atoms, for instance dimethyldodecylphosphine oxide; and dialkyl sulphoxides of structure R₂SO where the group R is an alkyl group of from 10 to 18 carbon atoms and the other is methyl or ethyl, for instance methyltetradecyl sulphoxide; fatty acid alkylolamides; alkylene oxide condensates of fatty acid alkylolamides and alkyl mercaptans.

Suitable amphoteric detergent-active compounds that optionally can be employed are derivatives of aliphatic secondary and tertiary amines containing an alkyl group of 8 to 18 carbon atoms and an aliphatic radical substituted by an anionic water-solubilizing group, for instance sodium 3-dodecylamino-propionate, sodium 3-dodecylaminopropane sulphonate and sodium N-2-hydroxydodecyl-N-methyltaurate. Suitable cationic detergent-active compounds are quaternary

ammonium salts having an aliphatic radical of from 8 to 18 carbon atoms, for instance cetyltrimethyl ammonium bromide.

Suitable zwitterionic detergent-active compounds that optionally can be employed are derivatives of aliphatic quaternary ammonium, sulphonium and phosphonium compounds having an aliphatic radical of from 8 to 18 carbon atoms and an aliphatic radical substituted by an anionic water-solubilising group, for instance 3-(N-N-dimethyl-N-hexadecylammonium) propane-1-sulphonate betaine, 3-(dodecylmethyl sulphonium) propane-1-sulphonate betaine and 3-(cetylmethylphosphonium) ethane sulphonate betaine.

Further examples of suitable detergent-active compounds are compounds commonly used as surface-active agents given in the well-known textbooks "Surface Active Agents", Volume I by Schwartz and Perry and "Surface Active Agents and Detergents", Volume II by Schwartz, Perry and Berch.

The detergency builders used in the formulation are preferably inorganic and suitable builders include, for example, alkali metal aluminosilicates (zeolites), alkali metal carbonate, sodium tripolyphosphate (STPP), tetrasodium pyrophosphate (TSPP), citrates, sodium nitrilotriacetate (NTA) and combinations of these. Builders are suitably used in an amount ranging from 1 to 50% by wt. Zeolite if used as builder is present at levels not exceeding 10% by wt.

THE KIT

The polymers of the invention are added to the detergent formulation using conventional methods of incorporation. The polymers can also be used as art of a fabric washing kit which is described.

The fabric washing kit has two enclosures separated from each other, one big and the other small. The big enclosure contains the detergent formulation and the small enclosure the polymer. An instruction sheet contains directions for the use of the two components including the proportions and conditions under which these are to be used. The kit may also contain a dispensing means to aid the washing using the two components in the desired proportions.

A typical kit according to the invention is a plastic container having two separate chambers. A first bigger chamber holds the detergent composition. The small chamber has a volume capacity of about 10% of the big chamber in which the polymer is provided. The instruction material is mounted on the side of the lid facing the container.

The polymer/detergent composition may also be packaged in single dose sachet having compartment in which the two components are housed in a manner such that they come into contact when the contents are discharged from the sachet. It is also possible to microencapsule the polymer and provide the product in a premixed form. Other forms of packagings are also included within the scope of invention.

The nature of the invention, its objects and advantages will be more apparent from the ensuing description made with relation to non-limiting exemplary examples of the above identified various aspects of the invention.

EXAMPLES.

a. Synthesis of soil release polymer:

100 g urea was dissolved in 1 litre of distilled water in a flask equipped with a stirrer and a thermometer. 1 kg of tapioca starch, 1 g ferrous ammonium sulphate and 5 g ascorbic acid, 50 ml. methyl acrylate were added sequentially and the mixture was stirred. 10 ml hydrogen peroxide (30% w/v) was then added, the reaction mixture stirred and then filtered. The reaction was conducted at 30°C. The hydrophobic graft copolymer of starch-graft-methyl acrylate obtained was repeatedly washed with water and then dried at 100°C.

250 g of chloroacetic acid and 240 g of sodium hydroxide was dissolved in water and mixed under stirring maintaining a temperature of <20°C. 40 g urea was then added to the mixture. 1 kg of the above starch-graft-methyl acrylate was taken in a mixer and the chloroacetic acid-sodium hydroxide mixture was then sprayed on to the polymer under stirring. The mixture was left for 24 hours at 60°C and then dried to 11% moisture.

b. Demonstration of soil removal properties of the polymer:

Detergent formulations were formulated with and without the incorporation of the polymer prepared as described above (Carboxylated Starch –g-Methyl acrylate) and the formulation details are presented in Table 1 and were tested for soil release properties which is determined by improvement in detergency.

Table 1

Composition % wt.	Example 1	Example 2
Linear alkyl benzene	20	20
sulphonate	,	
Sodium	27	27
tripolyphosphate		
Soda	15	15
Alkaline silicate	10	10
fillers	15	15
Carboxylated Starch -g-	-	2
Mėthyl acrylate	*	
Water	To 100	To 100

Test fabrics

100% Cotton and 100% polyester fabric were used in the study. The fabric was cut into swatches of dimension 5" X 5". The swatches were soiled using a) oily soil (Artificial sebum or Motor oil

+ oil soluble dye as an indicator) and b) particulate soil (carbon soot) c) commercially available fabric presoiled with a mixture of oily and particulate soil (WFK 30D was pre-soiled polyester and WFK 10D was pre-soiled cotton)

Soiling

0.2g of soil along with oil red dye at a concentration of 0.025% was loaded on to each of the test swatches and the stain was allowed to wick for a period of 24 hrs. Initial reflectance measurement at 520nm was taken, on a Milton Roy Color Scan II. R520 is the wavelength at which the red dye absorbs, hence it is used to monitor soil removal.

For particulate soil loading, carbon soot was deposited to the cloth piece to get a reflectance of 55 (at 460 nm).

Wash Conditions:

The fabric mentioned above were washed using the detergent compositions of Example 1 and 2 maintaining 10 replications for each.

Test swatches were washed in a tergo-to-meter in the detergent solution (Examples 1 and 2) for a period of 15 minutes. Reflectance measurements were taken at 520nm (for oily soil) or 460 nm (for particulate soil). The difference in reflectance of the soiled fabrics before and after washing was noted and represented as $\Delta R520*$ or $\Delta R460*$.

Improvement in oily soil removal ($\Delta\Delta R520^*$) =

 Δ R520* (for Example 2) - Δ R520* (for Example 1).

The improvement in soil removal for particulate and mixtures of oily and particulate was determined as follows:

Improvement in soil removal ($\Delta\Delta R460^*$)= $\Delta R460^*$ (for Example 2) - $\Delta R460^*$ (for Example 1)

The results of the tergo-to-meter washes using detergent solution with and without the polymer following the above detergency test procedure for removal of soils from polyester is presented in Table 2 and for cotton in Table 3..

Table 2

Detergency benefit on polyester fabric

Example	Particulate	Artificial Sebum	Motor Oil	Mixture of oily &
	soil		·	particulate soil
	ΔΔR460*	ΔΔR520*	ΔΔR520*	ΔΔR460*
Example 2	2.4	1.6	2.5	2

Table 3

Detergency benefit on cotton fabric

Polymer used	Particulate soil	Artificial Sebum	Motor Oil	Mixture of oily & particulate soil
	ΔΔ R 460*	ΔΔ R520 *	ΔΔ R520 *	ΔΔR460*
Example 2	3	0.5	0.5	2

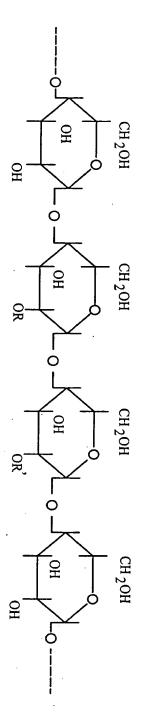
The above reflectance measurements indicate benefits in use of anionically modified hydrophobic graft copolymer of starch in removal of both oily and particulate soil or their mixture on both polyester and cotton fabric.

It has thus been possible to provide for novel polysaccharide polymers based on hydrophobic and anionic modification of polysaccharides and a process for making the same. Their benefit with respect to soil release from both polyester and cotton fabric has been significant in detergent formulations.

Dated this 9th day of May 2000.

HINDUSTAN LEVER LTD.

Patents Manager



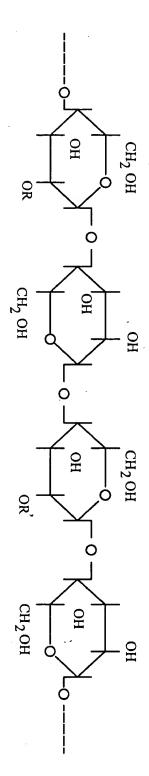
FORMULA II

FIG. 1a: ANIONIC HYDROPHOBIC GRAFT COPOLYMER OF STARCH

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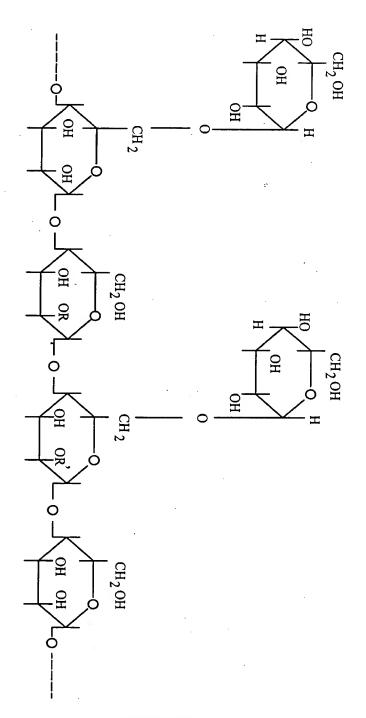
FORMULA II

FIG. 1b: ANIONIC HYDROPHOBIC GRAFT COPOLYMER OF CELLULOSE.

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FORUMLA II

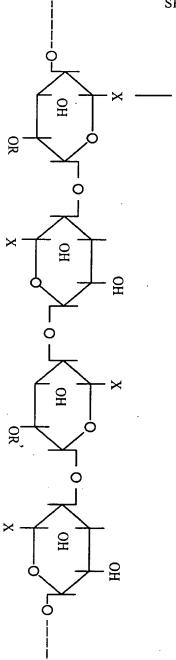
FIG. 1c: ANIONIC HYDROPHOBIC GRAFT COPOLYMER OF GUAR GUM.

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Where
$$X = H$$
 H
 OH
 OH



FORUMLA II

FIG. 1d: ANIONIC HYDROPHOBIC GRAFT COPOLYMER OF TAMARIND GUM.

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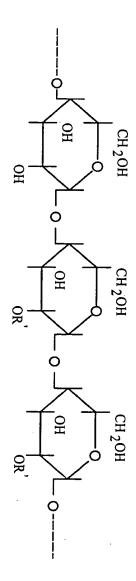


FIG. 2 a: ANIONIC MODIFICATION OF STARCH.

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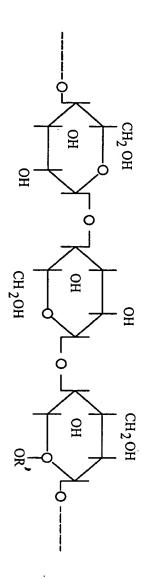


FIG. 2 b: ANIONIC MODIFICATION OF CELLULOSE

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: 1

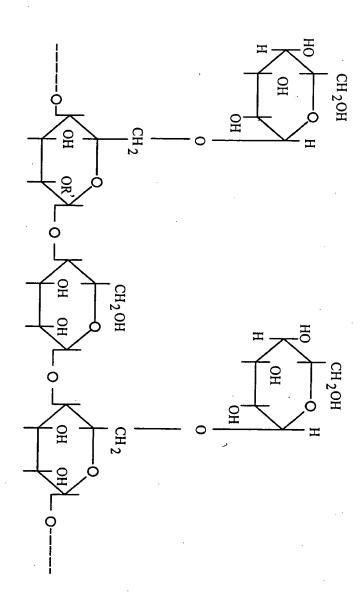


FIG. 2 c : ANIONIC MODIFICATION OF GUAR GUM.

Dated 9 th may 2000

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- 9 MAY 2000

FIG. 2 d: ANIONIC MODIFICATION OF TAMARIND GUM.

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(S Venkatramani) Patents Manager

- 9. MAY 2000

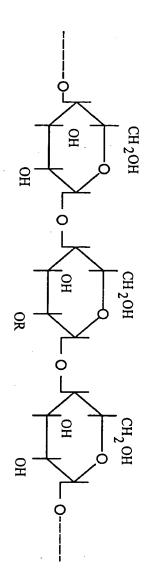


FIG. 3a: HYDROPHOBIC GRAFT COPOLYMER OF STARCH

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(S Venkatramani) Patents Manager

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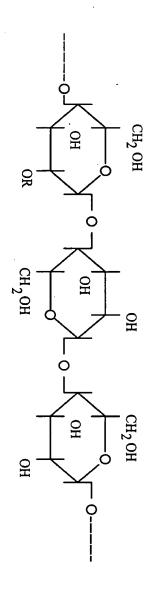


FIG. 3b: HYDROPHOBIC GRAFT COPOLYMER OF STARCH

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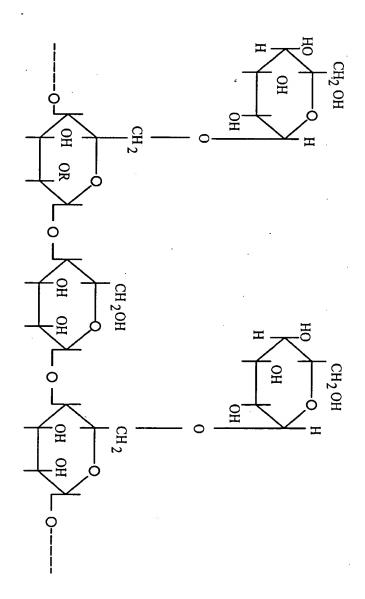


FIG. 3 c: HYDROPHOBIC GRAFT COPOLYMER OF GUAR GUM.

HINDUSTAN LEVER LIMITED.

Where
$$X = H$$

$$G = OH$$

$$GH_{OH}$$

FIG. 3 d: HYDROPHOBIC GRAFT COPOLYMER OF TAMARIND GUM.

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